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THE MECHANISMS OF FALLOUT PARTICLE FORMATION:
ANNUAL PROGRESS REPORT FOR PERIOD ENDING JUNE 1971

BALLISTIC RESEARCH LABORATORIES

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THE MECHANISMS OF FALLOUT PARTICLE
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by

Ralph F. Benck
Carl Crisco
Charles J. Mastrangelo
Stephen V. Pope
Alfonso W. Runquist

June 1973

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<p>This report was originally prepared in letter form and transmitted in June 1971 to the US Atomic Energy Commission, Division of Biology and Medicine, Fallout Studies Branch, Washington, D.C. under provisions of contract AT (49-7) 2883.</p>		

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VULNERABILITY LABORATORY

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SVPope/AWRunquist/tah
Aberdeen Proving Ground, MD
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I. INTRODUCTION

The research reported herein was undertaken by the US Army Ballistic Research Laboratories to study the basic phenomenology involved in the formation of radioactive fallout. Current efforts emphasize interactions between gaseous fission and activated elements and solid and liquid substrate materials. It is the purpose of this report to advise the sponsor of the program, the US Atomic Energy Commission, of the progress of the studies during FY 71.

This document does not include all the background material pertinent to each phase of the program. Only research areas which are new or drastically altered from previous reports are described in detail. Readers who may be interested in further specific details of the program are referred to progress reports for previous years¹⁻⁵.*

* References are listed on page 44.

II. SUMMARY

The accomplishments of this project during the period April 1970 - June 1971 are presented herein. This resume' is divided into five areas of study:

- (1) Sticking coefficient studies by the multiple collision method,
- (2) Sticking coefficient studies in the Molecular Beam Furnace, (3)
- Feasibility studies of sticking coefficient measurements on liquid (molten) substrates, (4) Electron microprobe analysis of fallout particles and (5) Desorption studies.

The major portion of the research effort was expended in areas 1, 2 and 3.

A. Significant Progress to Date

1. Several precise techniques have now been developed to study the attachment of fission product elements to various substrates. The agreement among the techniques is very satisfactory. An example of this agreement is illustrated in Table I which shows a comparison of results for silver on various substrates using techniques (1) and (2) above.

2. These results are significant in interpreting the radioactivity distribution observed in fallout particles. For instance, they confirm that it was not simply a peculiar accident that led to high levels of radioactivity in iron droplets in several real fallout particles which were studied in detail.

Table I. Sticking Coefficients of Silver at 20°C

Substrate	Sticking Coefficients	
	Multiple Collision Method ^a	Molecular Beam Method ^b
Silver	0.76	0.83
Iron	0.75	0.81
Quartz	c	0.54
Pyrex	0.68	0.60
Copper	0.71	0.71 ^d

^a Analyzed by comparison with Monte Carlo calculations of anticipated distributions for various sticking coefficients.

^b Analyzed by direct comparison of attachment to two different materials in otherwise duplicate experiments.

^c Not measured.

^d Assumed.

3. It is now possible to start to make general predictions regarding oxide and metal condensation behavior. Such generalizations are important because it is not feasible to study all fission elements condensing on all possible soil substrates.

4. For metals condensing on metals, a pattern is emerging:

(a) if both metals are "good" metals (with strong metallic resonance, i.e., copper, silver, or iron) the attachment will probably be high at all temperatures, at least up to 500°C, (b) If one metal is a "poor" metal (i.e., antimony or liquid tin), the condensation coefficient will be a little lower but not small.

5. For metals condensing on oxides, a very different pattern is followed. Several oxides have been tested and all show much similarity. There is fair attachment at low temperatures, but the metal sticking coefficient drops rapidly as the temperature is raised. At high (500°C) and very high (1500°C) temperatures the sticking coefficient is very low. This applies for acid quartz glass (SiO_2), for mixed acid-base but predominately acid Pyrex (80 weight per cent SiO_2), and for basic crystalline mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, only tested at 570°C).

6. Liquid metals are not necessarily excellent condensation surfaces. Molten tin, for example, rejects two thirds of the silver atoms which strike its surface under the experimental conditions used (which might involve an oxide film).

7. Vaporized soils can probably be treated as the individual oxide components (e.g., Al_2O_3 , K_2O , SiO_2) as shown by soil vaporization studies.⁶ Like compositions will behave similarly. Thus, SiO_2 (quartz glass) and 80 weight per cent SiO_2 (Pyrex glass) behave similarly if a correction factor is added for the borates present. Specifically, from 40°C to 250°C, the sticking coefficient of silver on Pyrex is approximately one-half that for quartz. At higher temperatures, the quartz value continues to fall and becomes even lower than the Pyrex value, but both are very low, i.e., 10^{-3} .

B. Multiple Collision Method of Sticking Coefficient Studies.

Studies of silver condensing on Pyrex have been terminated. The data have been very informative in showing the effect of substrate temperature by obtaining precise sticking coefficients at temperatures of ca. 20° to 250°C. The precision of the sticking coefficients, at temperatures greater than 250°C, is poor, but the data are still informative in that they indicate a pattern of behavior. The cause for the loss of precision may be that the assumptions necessary for the Monte Carlo calculations are no longer valid for silver condensing on Pyrex at temperatures exceeding ~ 250°C. These studies have been discontinued because further data would be of little additional value as well as being more difficult to obtain.

Studies of silver condensing on quartz substrates are also nearly complete. A few measurements at higher temperatures are required to confirm the condensation behavior as a function of temperature. A brief study of the mullite, $(3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2)$ - silver beam system has been conducted. The sticking coefficient of silver on mullite at 570°C is $\sim 6 \times 10^{-4}$.

Extensive studies on iron and iron oxide substrates had been planned but only two experiments were successful. The nonavailability of a suitable iron substrate was the primary difficulty, but with improved vacuum deposition techniques, this problem appears to be solved.

A technique has been developed to form an iron oxide substrate by passing steam through hot (500° to 600°C) iron coated glass or quartz cylinders.

The condensation of silver on silver, iron, and copper substrates has shown a different trend than that observed for an oxide substrate. The oxide sticking coefficients decreased with rising substrate temperature but the metal sticking coefficients are high at room temperature and increase with rising substrate temperature to a constant value.

Studies using antimony vapor were initiated. These studies should yield interesting results as the chemistry of antimony is similar to that of iodine and antimony is an important precursor in several of the fission chains.

C. Molecular Beam Studies

The data obtained using this technique are in good agreement with values determined by the multiple collision method. One series of measurements were performed with ^{110m}Ag on a quartz substrate at 1500°C. The low count rates on the substrate caused a low confidence level in the resultant sticking coefficient value (9×10^{-4}). A higher specific activity silver source would increase the confidence in the data, however, the results do demonstrate that the sticking of silver on quartz at 1500°C is very low.

D. Sticking Coefficient Studies on Liquid (Molten) Substrates

Two initial experiments with silver condensing on molten tin at 430°C indicated that the technique is feasible and with some minor refinements should be an excellent tool to study molten systems. Tin was chosen as the substrate for this study because of its low melting point and low vapor pressure. For future studies, gaseous fission products condensing on aluminum or on eutectics of uranium with iron or chromium should yield information pertinent to fallout studies.

E. Electron Microprobe Analysis of Fallout Particles

Electron microprobe x-ray analysis in conjunction with autoradiographs of 18 fallout particles show that metal inclusions generally contain high concentrations of the radioactive fission products compared to the surrounding silicate - aluminum - calcium matrices. In general, this result is also indicated by the sticking coefficient studies in that the sticking coefficient (at least in the case of ^{110m}Ag) is higher on metal substrates than on silicon oxide substrates.

The particles in this study were formed 10 to 15 years ago and the radioactivity levels are very low. It might be argued that fallout particles with higher levels of radioactivity should have been analyzed. However we were interested in studying fallout particles from various environments and the only particles available from a coral environment were from the 1956 Pacific Test Series.

There have been recent Plowshare tests conducted in Nevada but the only advantage particles from these tests would have over the old particles is that the autoradiographs would require less exposure time and they might exhibit better definition of the radioactive deposits. Analysis of particles from environments other than those already studied would certainly be of interest.

F. Desorption Studies

These studies were undertaken to obtain information about sticking coefficients via a series of simple experimental measurements. The theoretical interpretation and significance of the generated data are not clear⁷. Until such time that the relationships between the desorption rate and sticking coefficients are clarified, additional efforts will not be made.

III. MULTIPLE COLLISION METHOD OF STUDYING STICKING COEFFICIENTS

A. Procedures

The experimental apparatus for these studies and the data reduction technique are described elsewhere⁸. The pressure of the reaction chamber for all the results reported herein was from 1 to 4×10^{-5} torr. For the distributions performed at 20°C, a water cooled condenser-collector was used. For studies at higher temperatures, a resistance heater and copper heat spreader were used. The temperature profiles of the collector were carefully measured with thermocouples as a function of 5 levels of current through the substrate resistance heater. The results of these measurements are shown in Figure 1. The thermocouple probe was so constructed that the minimum distance to the source crucible at which a reading could be taken was 1 cm. To obtain an estimate of the wall temperature at the bottom of the column, the curves were extrapolated as is indicated by the dotted lines in Figure 1. Based on some earlier, incomplete temperature measurements, these extrapolations appear to be valid. The lower temperature at the bottom of the column is probably due to the column sitting on the water cooled RF condenser.

The source for the gaseous atoms was either $^{110m}\text{Ag}^*$ or $^{125}\text{Sb}^{**}$ electroplated onto a tungsten disk. The silver had a radiochemical purity of greater than 99 percent and a specific activity of 17.86 Ci per gram. The ^{125}Sb was carrier free with a radiochemical purity of greater than 99 percent.

* New England Nuclear, Boston, Mass.

** International Chemical and Nuclear Corporation, Pittsburgh, Pa.

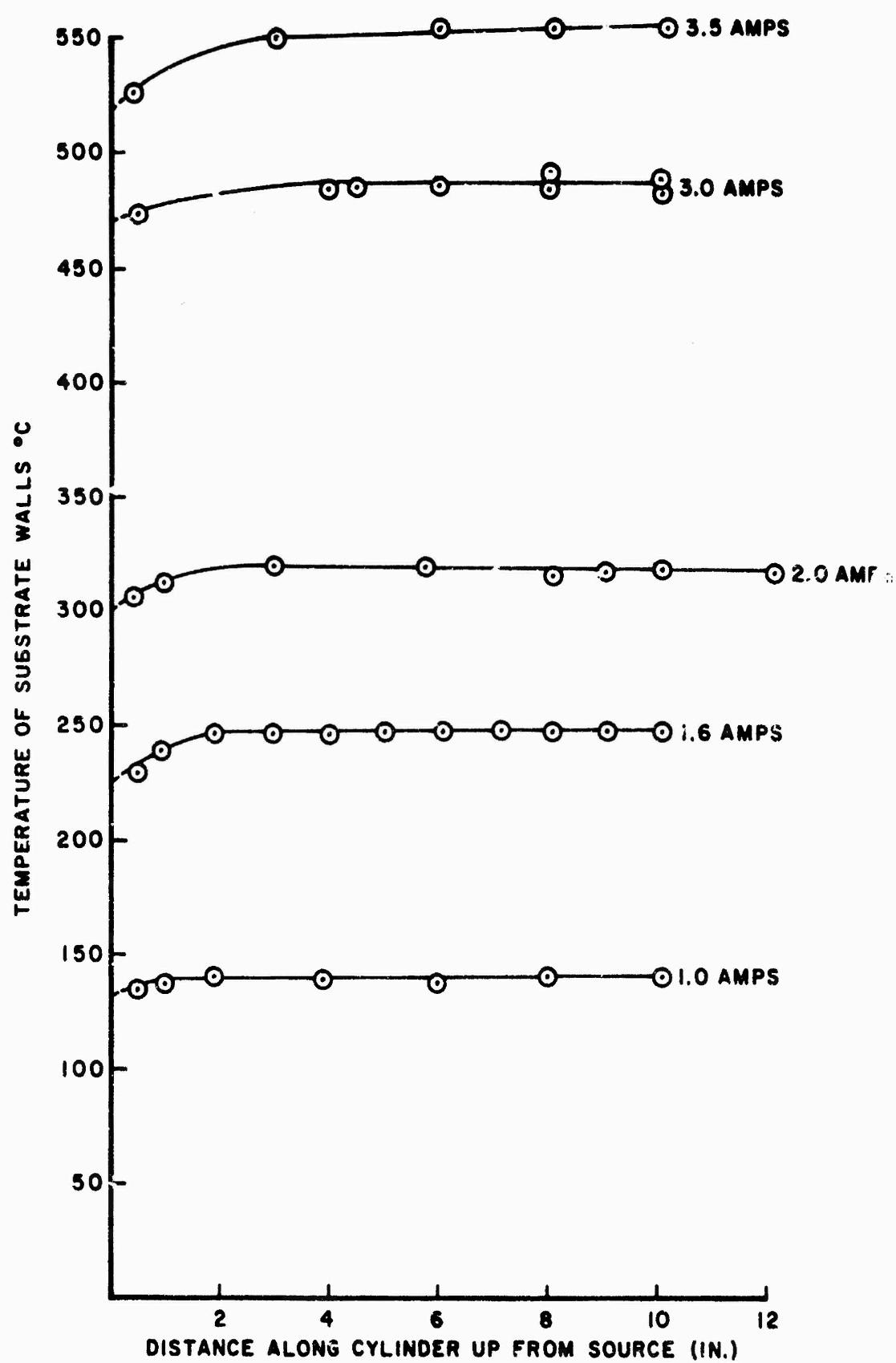


Figure 1 Temperature Profile of Substrate Walls.

The substrates used include Pyrex, quartz, silver, copper and iron. A single experiment was conducted with a $3\text{ Al}_2\text{O}_3\cdot\text{SiO}_2$ substrate (mullite tube*). Stock 2.54 cm. o.d. tubing was used for Pyrex and quartz substrates. Amorphous quartz glass was used for all the quartz substrates listed herein. The only experiments where crystalline quartz may have been used were those conducted at 1500°C. This transformation is discussed in a latter section of this report. For experiments with a heated copper substrate, a 0.38mm (15mil) piece of copper foil (99.9 percent copper as per electron microprobe analysis) was pressed against a copper heat spreader; for experiments at 20°C copper was vacuum deposited onto the inner surface of a Pyrex condenser.

It has been shown⁹ that basic properties of thin films are often quite dependent on film thickness and structure. It would have been desirable therefore to have used foil for all the copper substrate measurements.

A thin copper film was used at 20°C because the i.d. of the water cooled condenser, necessary to maintain 20°C, is such that only a very small thickness of copper could be used and still maintain a constant substrate diameter. For experiments at higher substrate temperatures, a heater was used that contained various thicknesses of copper heat spreaders. This arrangement allowed various thicknesses of copper foil substrates to be used while maintaining a constant substrate diameter.

The iron substrates were prepared by vacuum depositing high purity (99.999%**) iron onto the inner surface of 2.54 cm o.d. Pyrex tubing. Suitable iron foils were not available when the experiments with the heated iron substrate were performed. The silver substrates were prepared by silvering the inside surface of a Pyrex condenser of .54 cm o.d. Pyrex tubing (Brashear Process¹⁰).

The sticking coefficient is determined by measuring the amount of source material condensed on mica disks located above and below the cylinder openings. The mica was precoated with high purity (99.995%**) silver or antimony by vacuum deposition before being introduced into the reaction chamber.

* McDanel Refractories, Beaver Falls, Pennsylvania

** Electronic Space Products, Inc., Los Angeles, California.

The distribution of the condensed ^{110m}Ag or ^{125}Sb on the various substrates was determined by first cutting the substrates into 24, 1.27cm sections. Each section was then either leached with 5 N HNO_3 , and the dried residue beta counted, or gamma counted directly with a NaI scintillation crystal. With the ^{110m}Ag the experimental precision was approximately the same for both counting techniques. With ^{125}Sb , only direct counting was used as leaching was not quantitative.

B. Results

The sticking coefficients of silver on various substrates as a function of temperature are presented in Table II. All the results obtained to date by the multiple collision technique are included. The temperatures associated with some previously reported⁵ values have been changed to be consistent with an accurate temperature recalibration of the substrate walls. The averages of the sticking coefficient values in Table II are plotted versus the temperature of the various substrates and are shown in Figure 2.

The sticking coefficients of ^{110m}Ag and ^{125}Sb on Pyrex at 20°C are 0.68 ± 0.02 and 0.40 ± 0.07 respectively. These values are based on four condensations.

The deposition distribution of silver on the various substrates, generally approximates the Monte Carlo predicted distribution, e.g., Figure 3 for ^{110m}Ag condensing on Pyrex at 44°C and Figure 4 ^{110m}Ag condensing on copper at 485°C.

Monte Carlo predicted distributions of a material along a substrate are presented in Figure 5 for a sticking coefficient of 0.5, with and without specular reflections.

One deviation from ideality is demonstrated by the datum point for the top most section as shown in Figure 4. The abrupt increase in the amount of condensed ^{110m}Ag on this section has been observed for many experiments and may be due to the unique character or position of this section as it is the uppermost substrate section.

Table II. Sticking Coefficients of Silver On Various Substrates

Substrate Temperature °C	Pyrex	Silver	Copper	Iron	Quartz	$3Al_2O_3 \cdot 2SiO_2$ (Mullite)
20	0.68 ^a , 0.69	0.72, 0.79	0.67 ^b , 0.75	0.745 ^b , 0.75		
44	0.47 ^b					
65	0.43 ^b , 0.37 ^b , 0.37 ^b , 0.33 ^b	0.93	0.98 ^b		0.815 ^b	
105	0.20 ^b , 0.24					
145	0.215 ^b , 0.125 ^b , 0.14 ^b , 0.26 ^b	0.95	0.915 ^b		0.45	
235	0.04 ^b , 0.052 ^b , 0.048 ^b , 0.007 ^b , 0.0155	0.80, 0.81	0.89 ^b		0.54 ^b	
275	0.032 ^b					
320	0.048, 0.015 ^b	0.96, 0.87	0.88	0.91	0.16	
388			0.80 ^b			
485	0.04, 0.05, 0.15		0.92		0.011	
570					7×10^{-5}	6×10^{-4}

a Reported previously⁸b Previously reported⁵ but was incorrectly assigned to the wrong substrate temperature. The correct substrate temperature is now listed in column 1. See text.

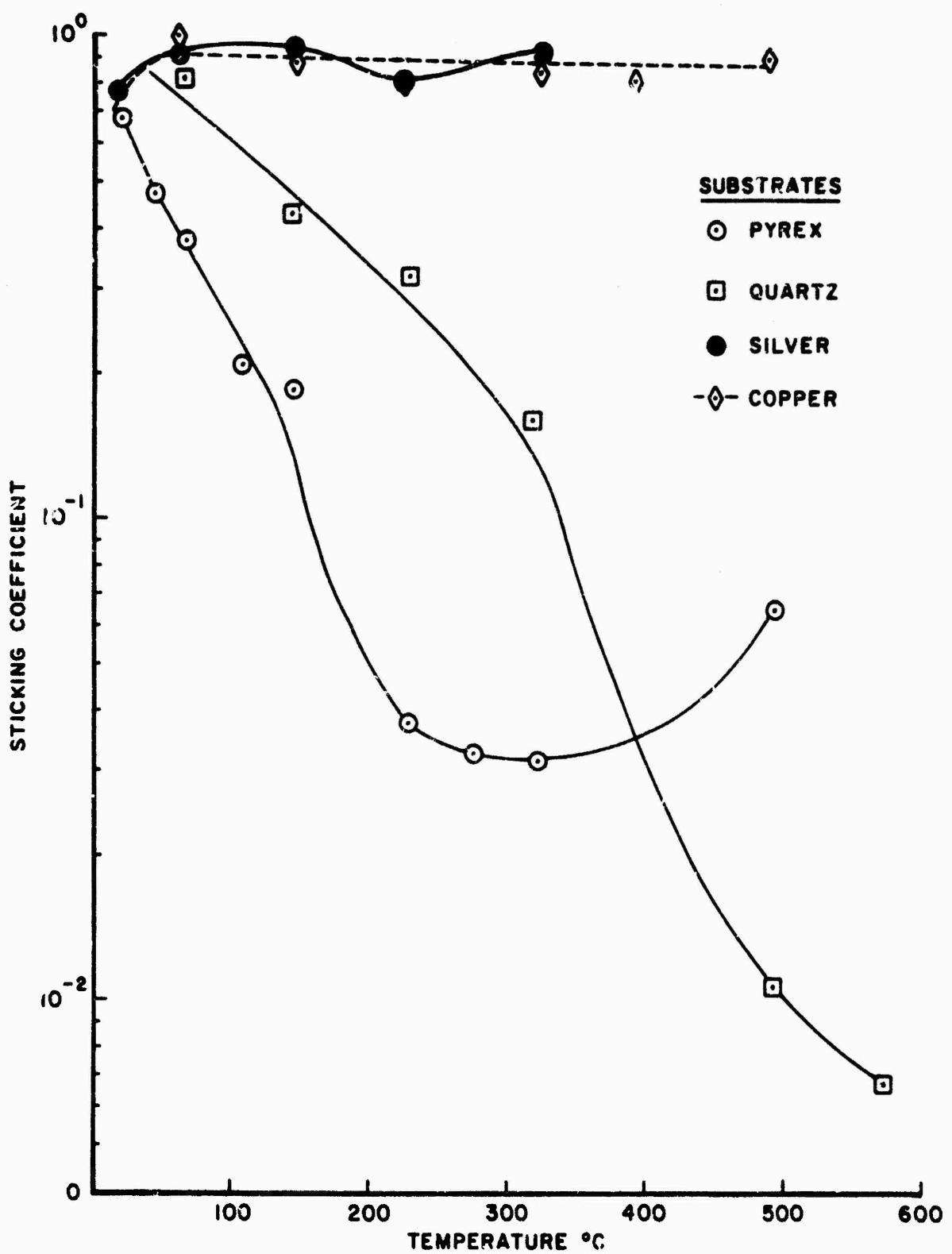


Figure 2 Sticking Coefficients of Silver as a Function of Temperature for Pyrex, Quartz, Silver and Copper Substrates.

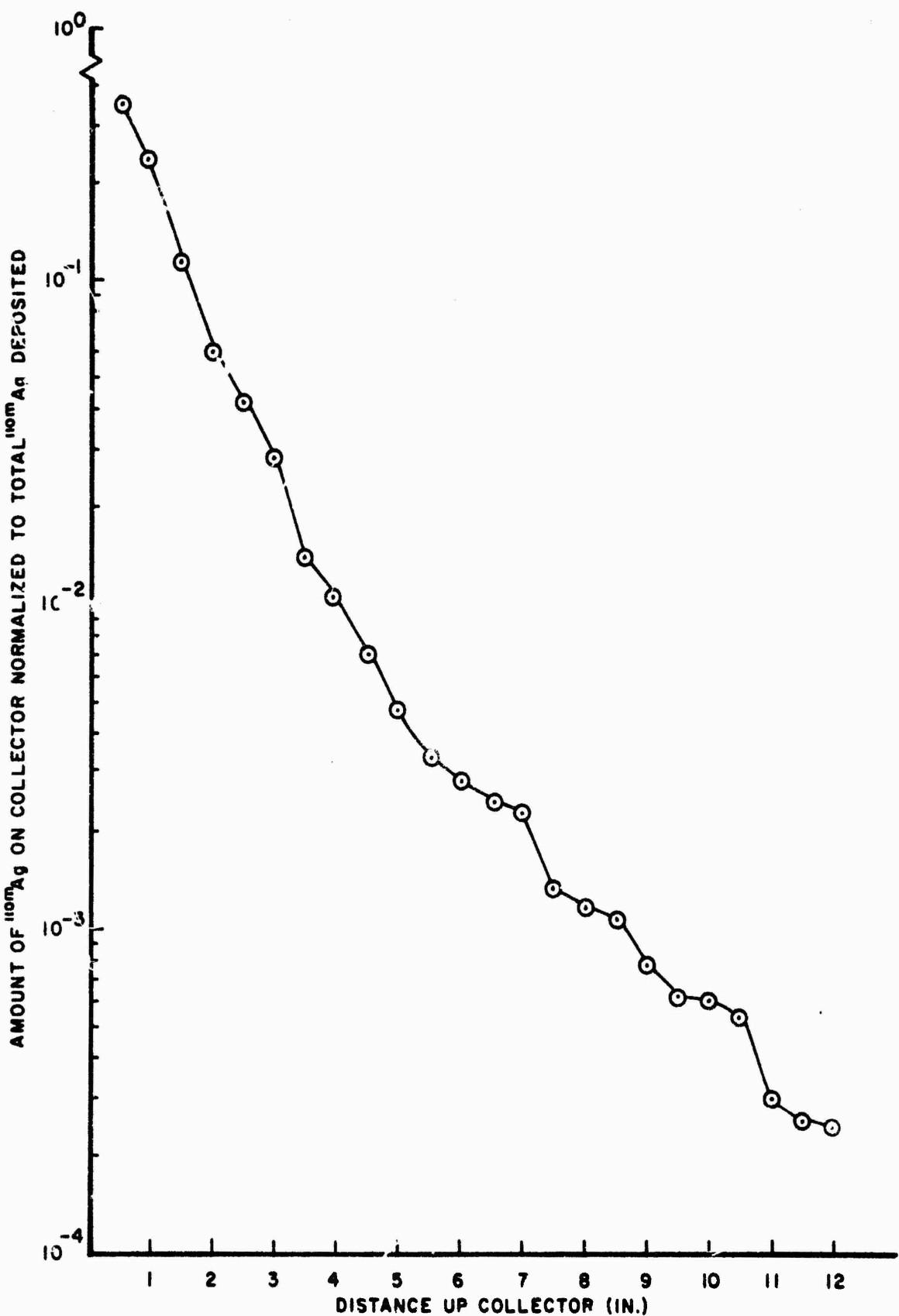


Figure 3 Distribution of Silver on a Pyrex Substrate at 44°C.

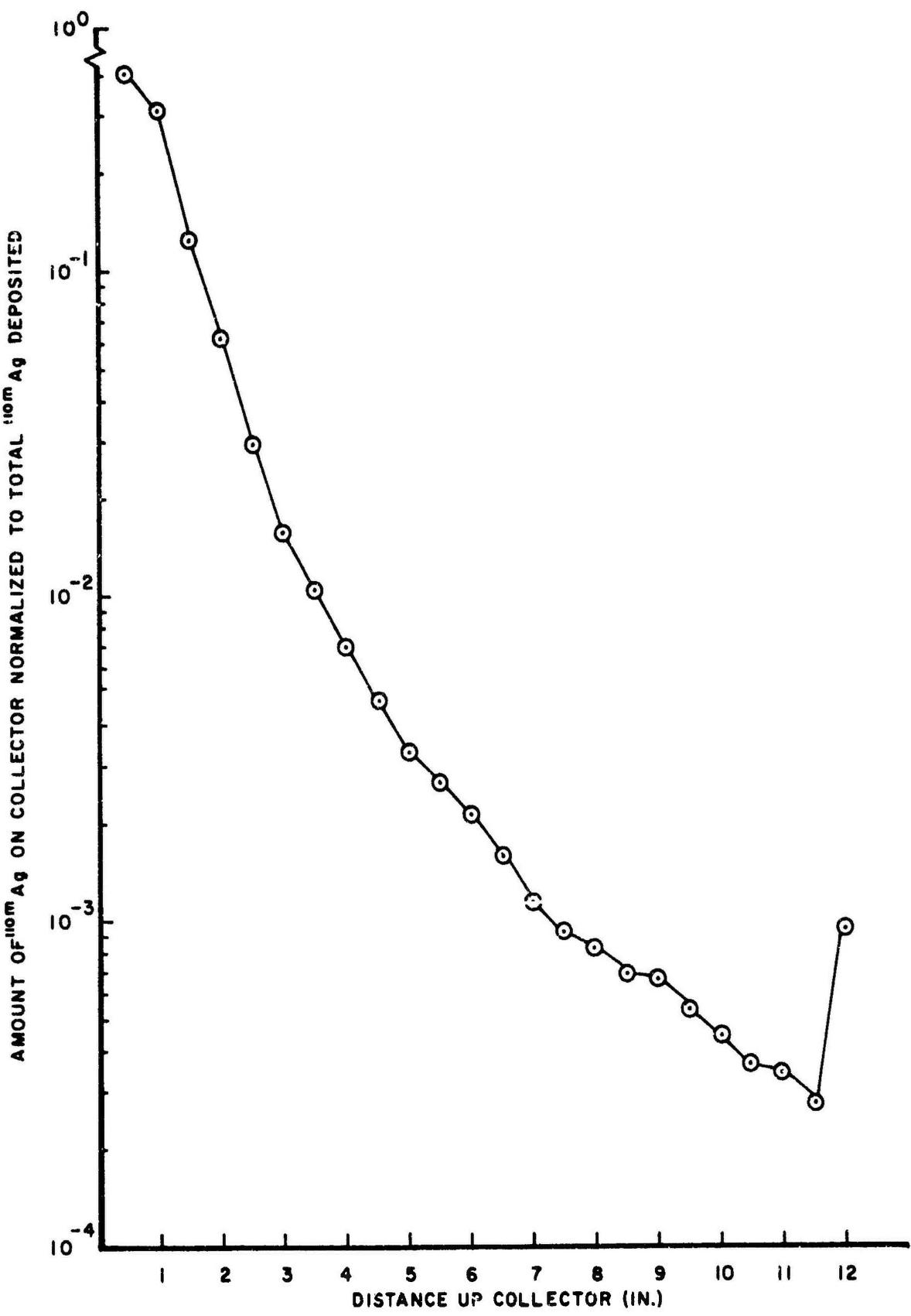


Figure 4 Distribution of Silver on Copper Substrate at 485°C.

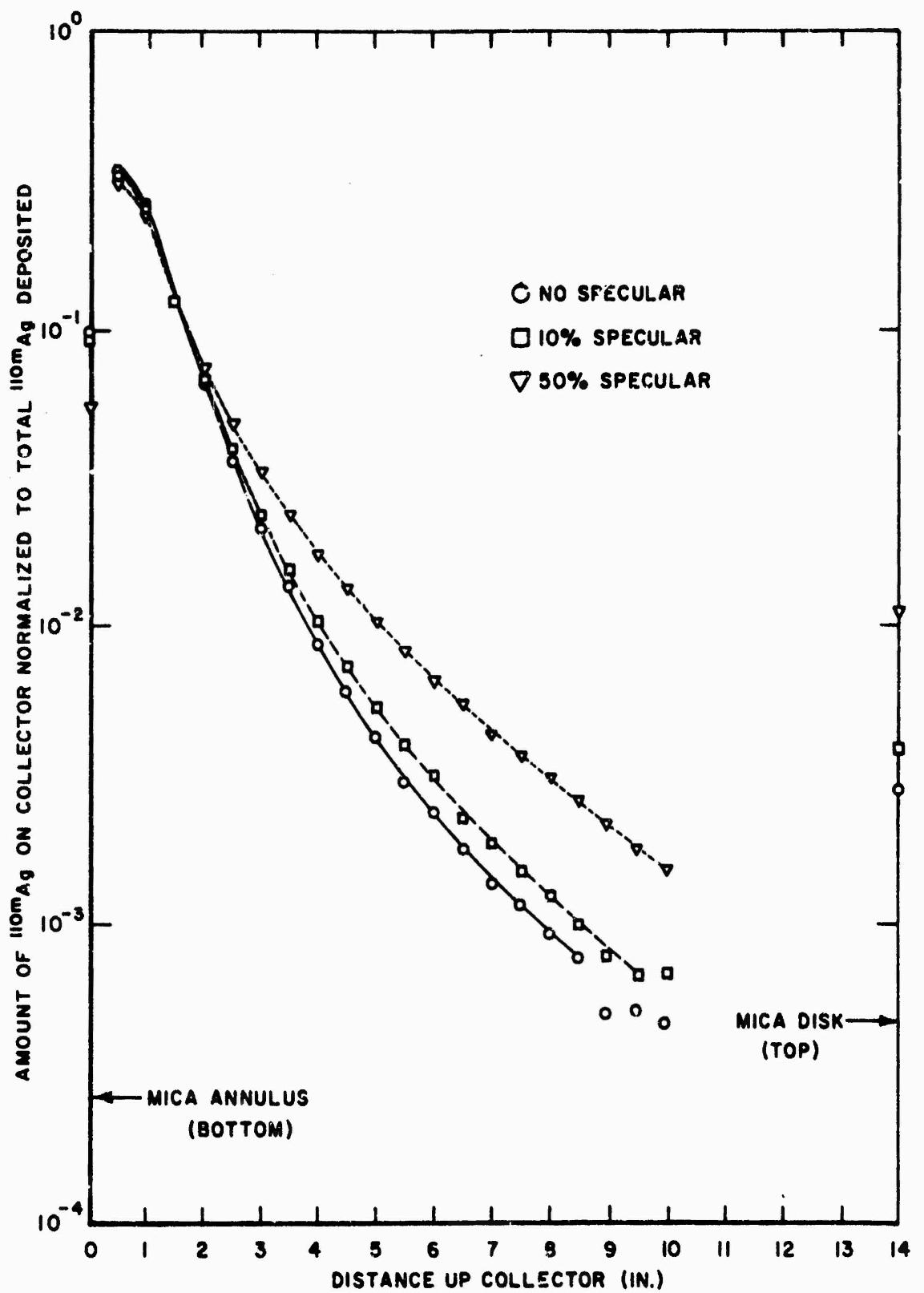


Figure 5 Theoretical Distributions as a Function of Percent Specular Reflections for a Sticking Coefficient of 0.5

The deposition distributions of ^{110m}Ag on Pyrex and quartz performed at substrate temperatures above 320°C are not ideal as indicated by the Monte Carlo curves. An example of this is shown in Figure 6 as the distribution of ^{110m}Ag on Pyrex at 485°C . Also, the shape of the distribution curve of ^{125}Sb on Pyrex at 20°C (Figure 7) differs from similar curves for ^{110m}Ag on Pyrex at 20°C .

C. Discussion

1. Silver Sticking on Pyrex and Quartz Substrates. The sticking coefficient of ^{110m}Ag condensing on Pyrex decreases with increasing substrate temperature to a minimum at approximately 300°C and then increases as indicated in Figure 2. One possible explanation for this increase in sticking coefficient with temperature is a change in the adsorption mechanism. Initially, the silver is physically adsorbed on the Pyrex surface the temperature of the Pyrex is increased, the adsorption mechanism changes from physical to chemical adsorption and an increase in the sticking coefficient occurs. The distribution of condensed silver on a 485°C Pyrex substrate, Figure 6, indicates that some mechanism in addition to diffuse reflections is taking place.

As is shown by the theoretical curve in Figure 5, specular reflections may be the additional factor in the condensation phenomenon. To fully evaluate the effects of specular reflections on the condensation distributions, theoretical curves, such as shown in Figure 5, should be determined for lower sticking coefficients.

Temperature gradients can be excluded as the additional factor. Figure 1 indicates that the temperature profile along the substrate is lower by approximately 15°C at the bottom but appears fairly uniform elsewhere. Since our studies have never indicated an abrupt change in the sticking coefficient over a temperature range of a few degrees, the possibility of the temperature causing the upward trend of the curves is not likely.

Other possibilities can be proposed, e.g., Pyrex surface softening, velocity shifts for the bouncing gas atoms, or competing effects by the

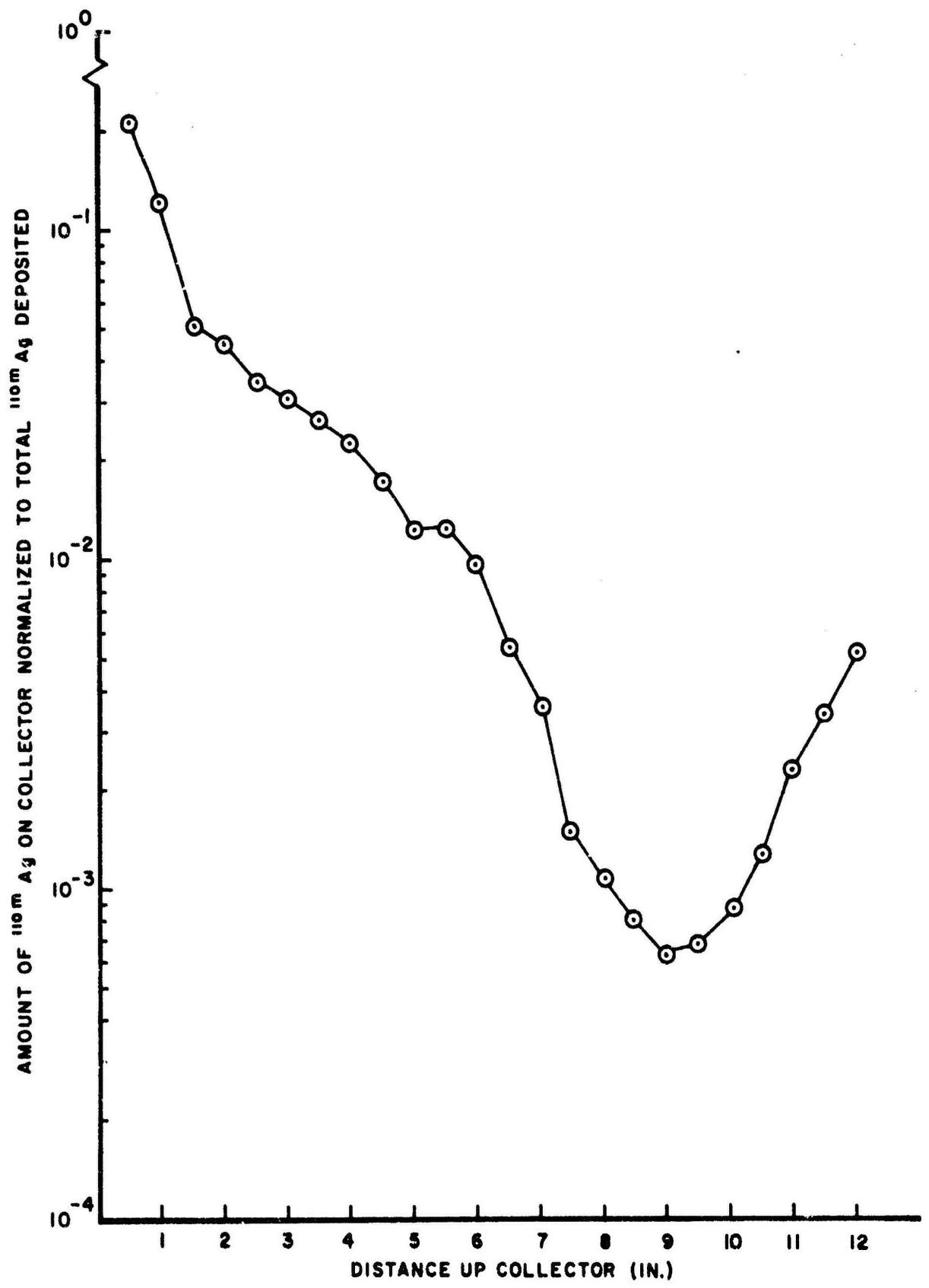


Figure 6 Distribution of Silver on Pyrex Substrate at 485°C .

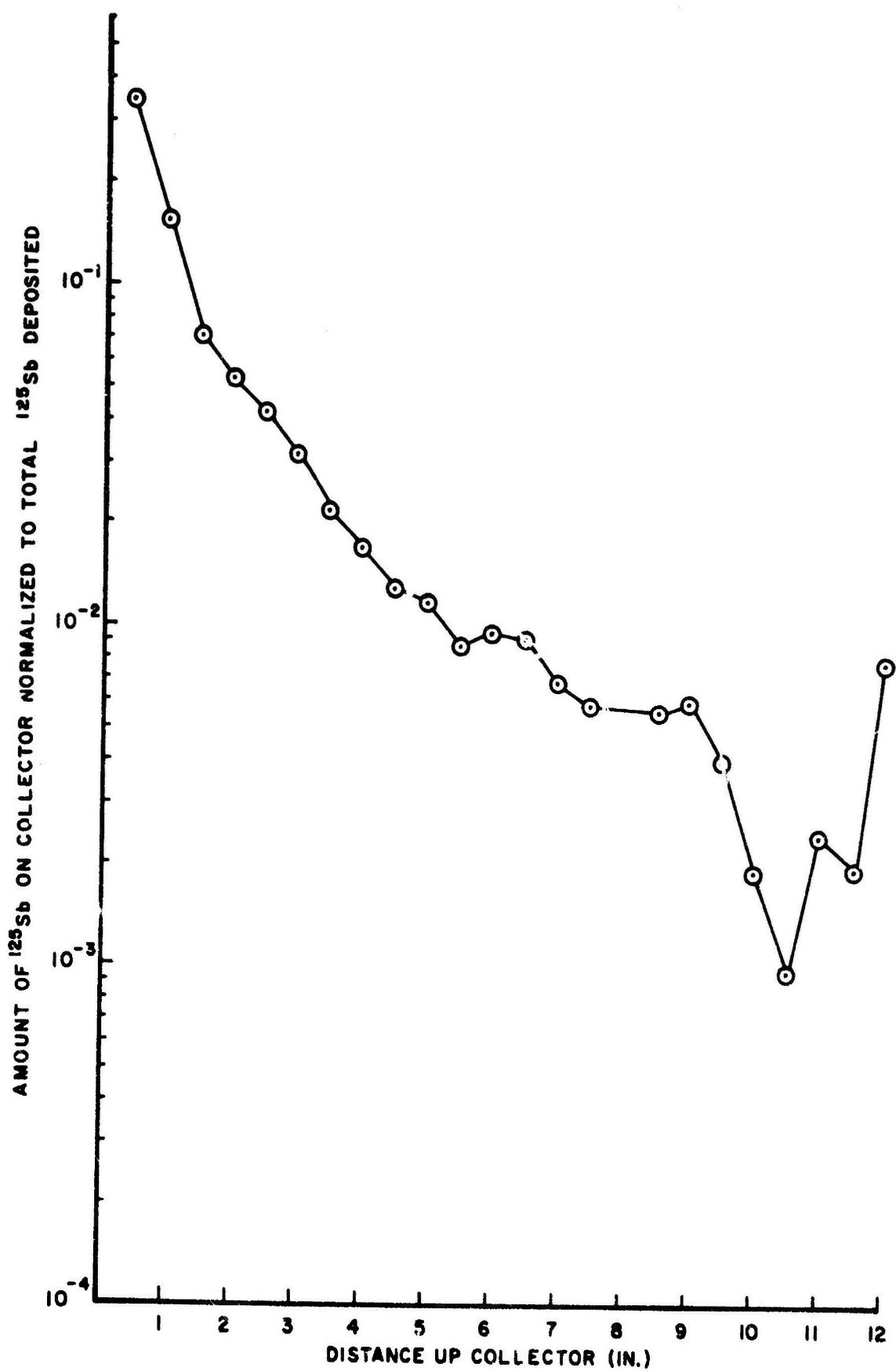


Figure 7 Distribution of Antimony on Pyrex at 20°C.

SiO_2 and B_2O_3 components of the Pyrex. For the present, it is not possible to choose among these and other possibilities. At any rate, the effect is small and is much reduced with pure SiO_2 (quartz glass).

The values of the sticking coefficients presented in Table II and Figure 2 are based on the amount of material adhering to an annular disk at the bottom of the column. As the sticking coefficient decreases, the relative amount of material condensed on the bottom disk increases, i.e., for a sticking coefficient of 0.01, with pure cosine reflections, 82 percent of the total evaporated gaseous material would be predicted by the Monte Carlo calculations to be condensed on the bottom disk. Therefore, due to the asymptotic nature of these curves, the lower limit of our present technique for pure cosine reflections is in the region of sticking coefficients between 5×10^{-3} to 5×10^{-4} . The lack of precision in the experimental values of silver on Pyrex at 485°C may be due to the values approaching this lower limit or due to appreciable specular reflections.

2. Condensation of Silver on Silver, Iron and Copper Substrates.

The sticking coefficient of ^{110m}Ag on copper, silver and iron is large but not unity at 20°C . This non-unity coefficient is probably caused by a surface contaminant such as water vapor^{11,12} present at this temperature and pressure. Heating of the silver and copper substrates to 65°C seems to degas some of this contaminant thus allowing the sticking coefficient above 65°C to rise to near unity. This apparent increase in sticking coefficient with temperature at high pressures has also been noticed previously in condensations of zinc on Pyrex¹³.

The sticking coefficient of ^{110m}Ag on copper is 0.9 up to the highest substrate temperature attained (485°C). There does not appear to be any problem in working with copper at higher temperatures. The experiments with ^{110m}Ag on substrates of iron and silver up to a temperature of 320°C also yielded a sticking coefficient of 0.9. In the case of silver and iron, however, the substrates have been vapor deposited thin films on Pyrex surfaces. For higher temperatures, the substrate support material will have to be changed to quartz.

It is a recognized fact that a thin film of metal often does not exhibit bulk properties of that metal⁹. Therefore, pure iron and silver foils should be used as a check against the possible anomalous behavior of thin coatings.

3. Condensation of Silver on Mullite. Only one experiment has been performed with the $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ Substrate. The measured value of the sticking coefficient of 6×10^{-4} is so low that the degree of confidence in the value is unknown. More measurements at this and lower temperatures should be obtained.

4. Condensation of ^{125}Sb on Pyrex at 20°C . The distribution curve of ^{125}Sb condensing on Pyrex (Figure 7), as mentioned above, is different than those for ^{110m}Ag condensing on Pyrex. The curve is similar to the curves shown in Figure 3 and may indicate some specular contribution to the distribution profile of the ^{125}Sb . The ^{125}Sb distribution curve also rapidly rises at the uppermost sections which is similar to that of ^{110m}Ag on Pyrex (Figure 6) except that the ^{110m}Ag starts to rise before the ^{125}Sb . The possible causes of this increase in activity in the upper sections should be similar to those mentioned in Section III C 1.

IV. MOLECULAR BEAM FURNACE

A. Procedures

A vacuum furnace has been fabricated to study the sticking coefficients of gaseous fission products on falling spheres at high temperature ($>1000^\circ\text{C}$). As mentioned previously⁵, due to shielding problems caused by the high levels of radioactive source material required, the original experimental plan has been altered and stationary flat substrates, rather than the falling spheres, have been used.

The present experiment utilizes a Knudsen Cell generated molecular beam of silver atoms impinging on stationary, flat substrates. The sticking coefficient of the silver on the flat substrates is determined by measuring the ratio of the amount of silver that adheres to the

amount that impinges on the substrates. Calibration of the silver flux was achieved by condensing the silver beam onto copper substrates at room temperature.

The pressure in the furnace for all the experiments described herein was in the 1 to 5×10^{-5} torr range. Studies using substrates of copper, silver, iron, Pyrex and quartz were performed at 20°C and quartz at 1500°C.

The copper (>99.9% as shown by electron microprobe analysis) was 0.38mm (15 mil) and the silver (99.999%*) was 0.13mm (5 mil). The iron substrates for the 20°C studies were prepared by vacuum deposition of several hundred Å of iron (99.99%**) onto cleaned pieces of copper foil. Once prepared, the iron coated samples were kept in a 0.1 mm vacuum, to minimize oxidation, until ready for use. The vacuum deposits of iron had to be used for the 20°C experiments because quality iron foils were not available at the time of experimentation.

For studies at 1500°C, pieces of 0.13 mm (5 mil) iron foil (99.99%**) were used. The Pyrex and quartz samples for the 20°C studies were polished, 2.54 cm diameter, 0.32 cm thick disks. The quartz samples for the 1500°C studies were 2.54 cm square, 1 mm thick, optically ground and polished quartz - CO grade ***. Different quartz substrates were used because the supply of the squares became exhausted and the disks were substituted. No differences between the quartzes were noted by either X-ray diffraction or electron microprobe analyses.

*Electronic Space Products, Los Angeles, California.

**Alpha Inorganics, Ventron, Beverly, Mass.

***ESCO Products, Oak Ridge, New Jersey.

All but three of the silver substrates were prepared in the following manner prior to being placed in the furnace: (1) wash in hot detergent* solution, (2) rinse in distilled water, (3) rinse in 2-propanol, and (4) dry in an oven at 125°C. Three of the silver substrates were put into the furnace in the "as received" condition.

The temperature of the Knudsen Cell was monitored during each experiment with an optical pyrometer**.

In the series of experiments of ^{110m}Ag condensing on quartz substrates at 1500°C, the substrate temperature was monitored by a Pt-Pt-10% Rh thermocouple placed adjacent to the quartz disks.

Initially there was difficulty in obtaining a constant silver beam. To circumvent this problem, a copper substrate was exposed along with the experimental substrate during each experiment. In this way the sticking coefficient, relative to copper, could be determined directly.

The samples were oriented normal to the ^{110m}Ag beam, 29cm from the Knudsen Cell, and situated such that the beam struck portions of both materials.

For experiments at 1500°C, single samples were exposed to the beam for 60 minutes. The amount of ^{110m}Ag deposited on the substrate was determined by counting the exposed samples in a low background (1 CPM), flow proportional, beta counter*** for 60 minutes or 10^5 counts. Quartz substrates were counted from 160 to 1080 minutes. The datum of each sample was converted to a deposition rate of atoms of silver per cm^2 of substrates surface per sec. The area exposed to the molecular beam was determined by measuring the area of the condensed ^{110m}Ag via a 1:1 autoradiograph of each substrate.

*Sparkleen, Fisher Scientific.

**Thermoscope-2 color pyrometer, Milletron, Inc., Pittsburgh, Pennsylvania.

***LOWBETA, Sharp Labs., Inc., La Jolla, California.

B. Results

The deposition rates of silver on the various substrates at 20°C are presented in Table III along with the average Knudsen Cell temperatures. The overall error, based on counting statistics, autoradiograph area and time of exposure uncertainties for each deposition rate is estimated to be \pm 3 percent.

The results of 7 exposures of quartz disks at 1500°C to the ^{110m}Ag beam are presented in Table IV. The temperature of the furnace and hence of the substrates was measured as $1500 \pm 8^\circ\text{C}$. The 0.13 mm (5 mil) iron substrates could not be used since they deformed at this temperature.

The deposition rate of the silver on the copper varied from (2.6 to 5.6) $\times 10^{11}$ atoms per cm^2 per sec (column 2 Table III) indicating that the molecular beam is reproducible to \pm 20 percent.

C. Discussion

Results in Section III indicate that under these experimental conditions the sticking coefficient of silver on copper, silver and iron is large but not unity at 20°C. As mentioned in section III this non-unity condensation at 20°C is probably caused by a surface contaminant such as water vapor present at this temperature and pressure.

The value of the sticking coefficient of ^{110m}Ag on copper at 20°C as described in Section III is 0.71. Using this value the following ratios can be used to determine the sticking coefficient of ^{110m}Ag on other substrates.

$$\frac{\text{Deposition Rate of}}{\text{---}} \frac{\text{---}}{\text{Deposition Rate of}} \frac{\text{Sticking Coefficient of}}{\text{---}} \frac{\text{---}}{\text{Sticking Coefficient of}} \\ \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \\ \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}}$$

=

$$\frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}} \frac{\text{---}}{\text{---}}$$

Ag on M

$\text{Ag on Cu (.71 at } 20^\circ\text{C)}$

where M is silver, Pyrex, iron or quartz. The ratios of the deposition rates of ^{110m}Ag onto the various substrates and the sticking coefficients as calculated from these ratios are presented in Table V.

Table III. Silver Deposition Rates at 20°C.

Experiment	Deposition Rate on Substrates of					Cell ^a Temperature °C
	Copper	Silver	Iron	Quartz	Pyrex	
Multiply all numbers by 10^{11} atoms $\text{cm}^{-2} \text{s}^{-1}$						
1	3.29	3.00 ^b				980
2	2.58	2.65 ^b				1001
3	3.44	2.57 ^b				994±3
4	4.04		4.75			1004±6
5	5.48		6.47			1018±3
6	5.03		5.38			1017±2
7	3.09				2.35	1010
8	4.27				3.87	1022±2
9	5.57				4.96	1026±1
10	3.04			2.17		1025
11	4.12			3.32		1021±4
12	4.74			3.63		1002±2
13	4.87	6.09				1022±2
14	2.65	2.91				1025±5
Avg.	4.01	3.44	5.53	3.04	3.73	
		(4.50) ^c				

^a The ± values are the limits observed in cell temperature fluctuation.^b Not cleaned prior to deposition (see text).^c Average, if first 3 values are not used.

Table IV. Silver Deposition Rates on Quartz Substrates at 1500°C

Experiment	^{110m}Ag Activity on Sample ^a Dis. min^{-1}	Deposition Rate ^a $10^8 \text{ atoms cm}^{-2} \text{ s}^{-1}$	Average ^b Knudsen Cell Temperature °C
1	17.0 ± 1.0	14 ± 0.8	1032 ± 12
2	4.9 ± 0.8	3.2 ± 0.5	964 ± 14
3	0.2 ± 0.4	0.1 ± 2.6	1013 ± 3
4	0.0 ± 0.4	0	1021 ± 6
5	4.9 ± 0.6	3.5 ± 0.4	1024 ± 4
6	0.2 ± 0.5	0.1 ± 3.0	1023 ± 3
7	20.3 ± 1.5	14 ± 1.0	1021 ± 20

^a Uncertainty based on counting statistics.

^b Limits of the temperature fluctuation of the cell.

Table V. Sticking Coefficients of Silver on
Silver, Iron, Quartz and Pyrex at 20°C

Substrate	Deposition Rate on Substrate M Divided by Deposition Rate on Cu ^a	Average of Experiments	Sticking Coefficient
Silver	1.17 ^b	13 & 14	0.83 ^c
Iron	1.14	4, 5, 6	0.81
Quartz	0.76	7, 8, 9	0.54
Pyrex	0.85	10, 11, 12	0.60

^a The results reported below are based on the specific experiments listed and not on the average deposition rate of silver on copper for the entire 14 experiments.

^b The first 3 silver runs (1, 2 & 3) have been discarded since the surface had not been cleaned and the cell temperature was approximately 20 degrees lower than in the other experiments.

^c If the first three runs are included, the sticking coefficient would be 0.71.

To evaluate the sticking coefficient of silver on quartz at 1500°C, the flux of silver atoms from the Knudsen Cell was taken to be the average of the amount of silver condensed on the 14 copper substrates at 20°C (Table III) divided by the sticking coefficient of ^{110m}Ag on copper at 20°C (0.71) or, $\frac{4.01 \times 10^{11} \text{ atoms cm}^{-2} \text{ s}^{-1}}{0.71} = 5.7 \times 10^{11} \text{ atoms cm}^{-2} \text{ s}^{-1}$.

The sticking coefficient of ^{110m}Ag on quartz at 1500°C would therefore be equal to:

$$\frac{\text{Deposition Rate at } 1500^\circ\text{C}}{\text{Flux Rate measured at } 20^\circ \text{ (} 5.7 \times 10^{11} \text{ atoms cm}^{-2} \text{ s}^{-1} \text{)}}$$

As seen in Table IV, the activity levels of the condensed silver on the quartz disks were above background but were low. Even long counting times resulted in poor counting statistics. Thus, large uncertainties exist in results from calculations based on these data. Keeping this in mind, the sticking coefficient of ^{110m}Ag on quartz at 1500°C ranges from 2×10^{-3} to 2×10^{-5} with the average being 9×10^{-4} which in this particular case approaches the limit of sensitivity.

V. STICKING COEFFICIENT MEASUREMENTS OF LIQUID (MOLTEN) SYSTEMS

If sticking coefficient measurements are to be conducted at substrate conditions close to those at which fallout is thought to form, a technique to measure sticking coefficients on molten substrates should be developed. The following discusses the feasibility of such a technique.

A. Procedure

An experimental apparatus to study the sticking coefficients of molten systems has been fabricated and is illustrated in Figure 8. It consists of a heated crucible containing the molten substrate and a molybdenum ribbon heater directly above the crucible. The molybdenum ribbon, bent in the form of a U, has the desired fission product source plated onto a given area of the surface facing the crucible. The relative amount of evaporated material from the molybdenum ribbon that impinges onto the molten substrate is reproducible due to rigid

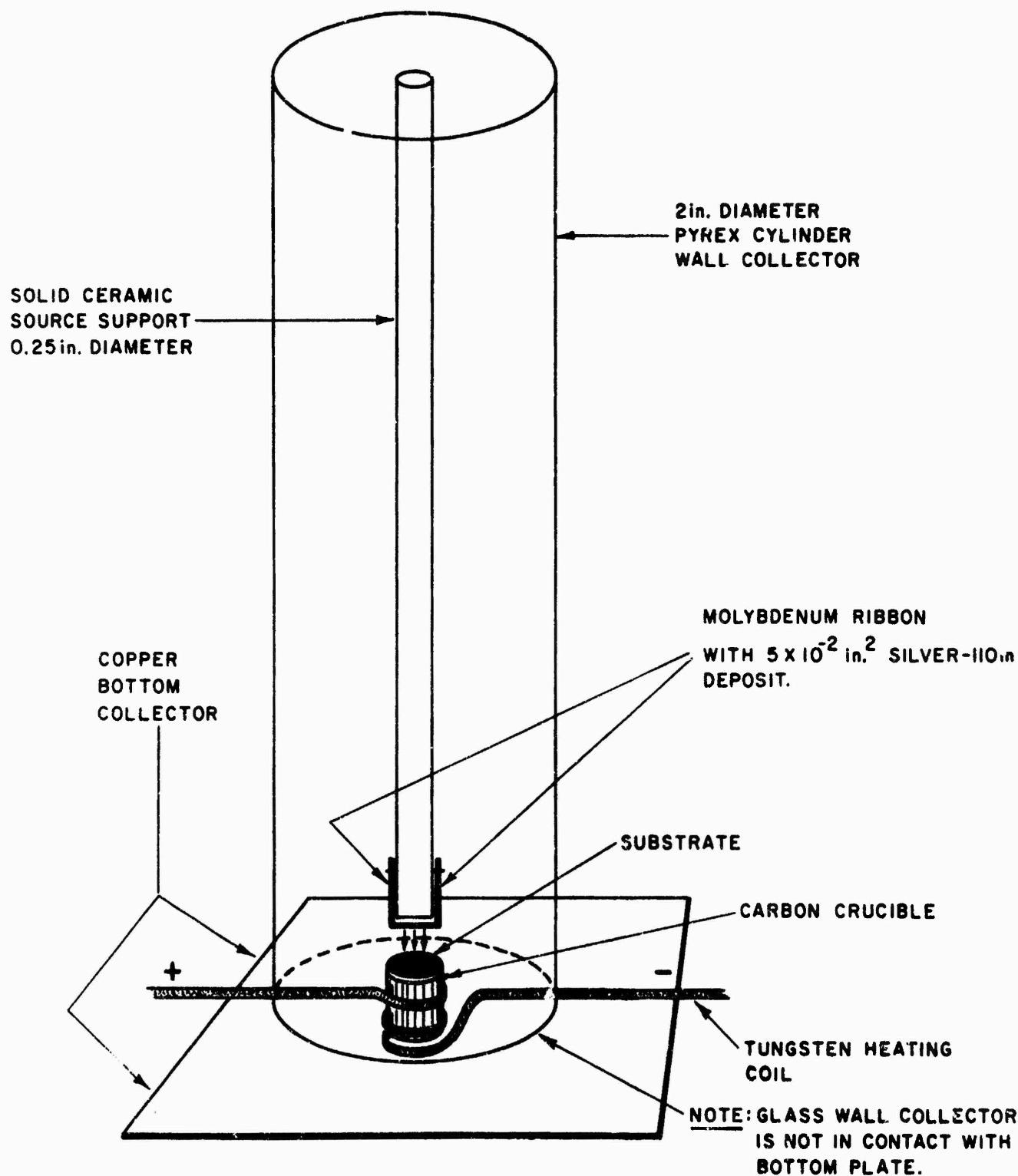


Figure 8 Sketch of Apparatus Used for Measurements of Sticking Coefficients on Molten Substrates.

control of the geometry of each experiment. Therefore, by measuring the amount of ^{110m}Ag evaporated from the molybdenum ribbon and the amount of ^{110m}Ag dissolved in the molten sample, the sticking coefficient of ^{110m}Ag in the molten sample can be calculated:

$$\text{Sticking Coefficient} = \frac{\text{Amount of } ^{110m}\text{Ag in Substrate}}{\text{Amount of } ^{110m}\text{Ag that should have struck substrate}}$$

For the results reported herein the following experimental conditions existed: (1) Crucible - carbon - 1.27 cm o.d., 1 cm i.d., 1.27 cm high, (2) Substrate-tin, National Bureau of Standards, freezing point standard, (3) Source to substrate distance, 1.6 cm, (4) Source, electroplated ^{110m}Ag , $0.3 \text{ cm}^2 (5 \times 10^{-2} \text{ in}^2)$ deposit, (5) Substrate temperature $430^\circ \pm 5^\circ\text{C}$, (6) Pressure - $2 \pm 1 \times 10^{-4}$ torr and (7) Wall Collector - Pyrex cylinder, 5.1 cm diameter 15 cm high. The crucible was heated to a constant temperature for 10 minutes. The overhead molybdenum heater was then heated rapidly to a constant temperature by passing current through it for approximately one minute. The cooled solid substrate was then assayed by either direct gamma counting or by beta counting a dried aliquot of the dissolved sample.

The amount of ^{110m}Ag that impinged on the molten sample was precalibrated by inserting a 10x12 cm, flat, copper plate at the bottom of the 5.1 cm cylinder to collect the ^{110m}Ag evaporated from the heater ribbon. Silver was evaporated as outlined above and the distribution on the copper and the Pyrex wall collector was assayed by beta counting. The ratio of the ^{110m}Ag atoms that intersected the crucible area to the total flux from the heater was therefore determined. The Pyrex wall collector was used to collect the silver not condensed on the copper plate. The amount of silver on the copper plate and on the Pyrex should add up to the amount of silver evaporated.

B. Results

The results of the ^{110m}Ag calibration experiment are shown in Table VI. The area listed as "Crucible Area" is that area which the molten substrate would occupy during an actual experiment. Table VII lists the

Table VI. Calibration of Molten Substrate Apparatus

A. Initial ^{110m}Ag on Heater	31,917	CPM
B. Final ^{110m}Ag on Heater	4,271	CPM
C. ^{110m}Ag Evaporated (A-B)	27,646	CPM
D. ^{110m}Ag Condensed on Crucible Area	2,733	CPM
E. ^{110m}Ag Condensed on Cu Plate (except Crucible Area)	11,917	CPM
F. ^{110m}Ag on Pyrex Wall Collector	12,920	CPM
G. Sticking Coefficient of ^{110m}Ag on Cu at 20°C^a	0.71	
H. Total ^{110m}Ag that intersected Crucible Area	3,850	CPM
I. Percent of total evaporated ^{110m}Ag that hit Crucible Area (H/C)	14.	%

^a -See Section II of this report

Table VII. Sticking Coefficients of ^{110m}Ag on Tin at 430°C

	Run No. 1	Run No. 2
A. Initial ^{110m}Ag on Mo Ribbon	CPM, 153,816	51,894
B. Final ^{110m}Ag on Mo Ribbon	CPM, 51,894	33,049
C. ^{110m}Ag Evaporated (A-B)	CPM, 101,922	18,845
D. ^{110m}Ag Condensed in Tin Substrate	CPM, 4,752	976.
E. ^{110m}Ag initially struck Tin (0.14xC)	CPM, 14,269	2,638
F. Sticking Coefficient of ^{110m}Ag on Tin at 430°C (D/E)	0.33	0.37

* Counts per minute

results of two experiments of ^{110m}Ag on molten tin at 430°C .

C. Discussion

The most important factor in calculating the sticking coefficient is that the percent of the total evaporated ^{110m}Ag that hit the crucible area (item I in Table VI) be reproducible. To determine item I it was necessary to include the non-unity sticking coefficient of ^{110m}Ag on copper at 20°C . In section III the sticking coefficient of silver on copper at 20°C was measured to be 0.71. This is the value that has been used in calculating the sticking coefficients of silver on molten tin as shown in Table VII. In section III the pressures used were approximately 5×10^{-5} torr which are lower than the $1-2 \times 10^{-4}$ torr in the molten substrate studies. Therefore 0.71 may be in slight error but is the best value available for this study.

Another factor to consider in these studies is that the molten tin meniscus causes the sample to be slightly dome shaped. If there is an incident angle dependence of the sticking coefficient, it is not accounted for in the experiment.

The results in Tables VI and VII are based on only one flux calibration and therefore the accuracy of the results is hard to estimate. The reproducibility is approximately 6 percent. A literature value for the sticking coefficient of silver on tin at 430°C could not be found so a comparison with other results could not be made. Tin was chosen as the substrate because it has a low melting point and low vapor pressure and any heating effects would be minimal. Once the technique is perfected, substrates such as lead and eutectics of carbon-iron and uranium could be used as the substrates.

D. Conclusions

The technique, as outlined above, appears to be a promising method to measure sticking coefficients on molten substrates.

VI. ELECTRON MICROPROBE ANALYSIS OF FALLOUT PARTICLES

A total of eighteen fallout particles consisting of 6 from operation Red Wing, 5 from Shot Sedan and 7 from Shot Johnnie Boy were analyzed via physical property characterization and electron microprobe X-ray analysis. The fallout particles were cut, sectioned and polished, and the radioactivity on the polished surfaces mapped by autoradiography. Qualitative and semi-quantitative analyses were performed on those areas delineated by the autoradiographs.

A comprehensive report on these in depth analyses plus the analyses of 9 particles reported previously⁵, is presently in publication and will be published as a BRL report. The results of qualitative analyses of the 18 particles are shown in Table VIII. A summary of the conclusions is as follows:

1. The Red Wing samples were small and fairly homogeneous. The Sedan and Johnnie Boy samples were larger and more heterogeneous. The major element present in Red Wing samples was calcium; the Sedan and Johnnie Boy samples consisted of a matrix of either silicon or aluminum with small amounts of calcium.

2. Semi-quantitative analyses of the fallout particles from Shots Sedan and Johnnie Boy (Nevada) showed that generally the largest amounts of radioactivity were associated with iron inclusions or deposits in the particles. Areas of particles that were low in iron content generally did not exhibit any radioactivity. In the particles from operation Red Wing (Pacific), generally iron was not detected, the radioactivity was concentrated in areas high in magnesium or high in crystallized sodium chloride. Some particles were similar to coral fragments (mainly calcium). These particles had radioactivity distributed evenly throughout.

Table VIII. Electron Microprobe Qualitative Analysis of Fallout Particles

Red Wing Samples	Elements Detected by Microprobe										
	Ca	Al	Fe	Mg	Si	Cl	Na	K	Mn	Zn	Ti
1	X										
2	X			X		X	X				
3	X										
4	X	X	X	X	X	X	X	X			
5		X	X			X	X				
6	X			X		X	X	X			
Sedan Samples											
51	X	X	X	X	X			X	X		
52	X	X	X		X			X		X	
53	X	X	X		X			X	X		
54	X	X	X		X			X			
55		X	X		X			X			
Johnnie Boy *											
Samples	X	X	X		X		X				

* Each of the 7 particles from this shot exhibited this elemental composition. However within each sample there was considerable heterogeneity.

VII. DESORPTION STUDIES

In an effort to study sticking coefficients at higher temperatures, the rate of desorption of a thin film from a substrate was measured for several metal-substrate pairs. Frauenfelder¹⁴ has indicated that desorption of a thin film from a surface was directly related to the sticking phenomena. If this conclusion is valid, desorption studies would be germane to sticking phenomena experiments.

The substrates used for these studies were disks 0.6 cm diameter by 0.08 cm thick. The thin films of silver and antimony on tungsten substrate were prepared by electrodeposition; the silver thin films on Pyrex substrates were prepared by vacuum deposition. The substrate disks were held in a copper holder and placed into a heated vacuum furnace at 4×10^{-4} torr for specific lengths of time.

Using these disks, the evaporation rates of silver and antimony from tungsten and of silver from Pyrex were studied. Desorption rates of the samples were determined by beta counting the samples before and after specified lengths of heating.

The results of these studies are plotted in Figure 9 as the atoms on the substrate versus the time that the substrates were at constant temperature.

One conclusion that can be drawn from Figure 9 is that it is easier to evaporate a thin film of silver from a tungsten surface at 700°C than it is to evaporate antimony from the same surface. This probably indicates a higher binding energy between antimony and tungsten than silver and tungsten at 700°C. The higher binding energy most likely indicates a higher sticking coefficient.

However, it was not possible to correlate the limited data in Figure 9 with any available sticking coefficient data. No further work in this area is contemplated.

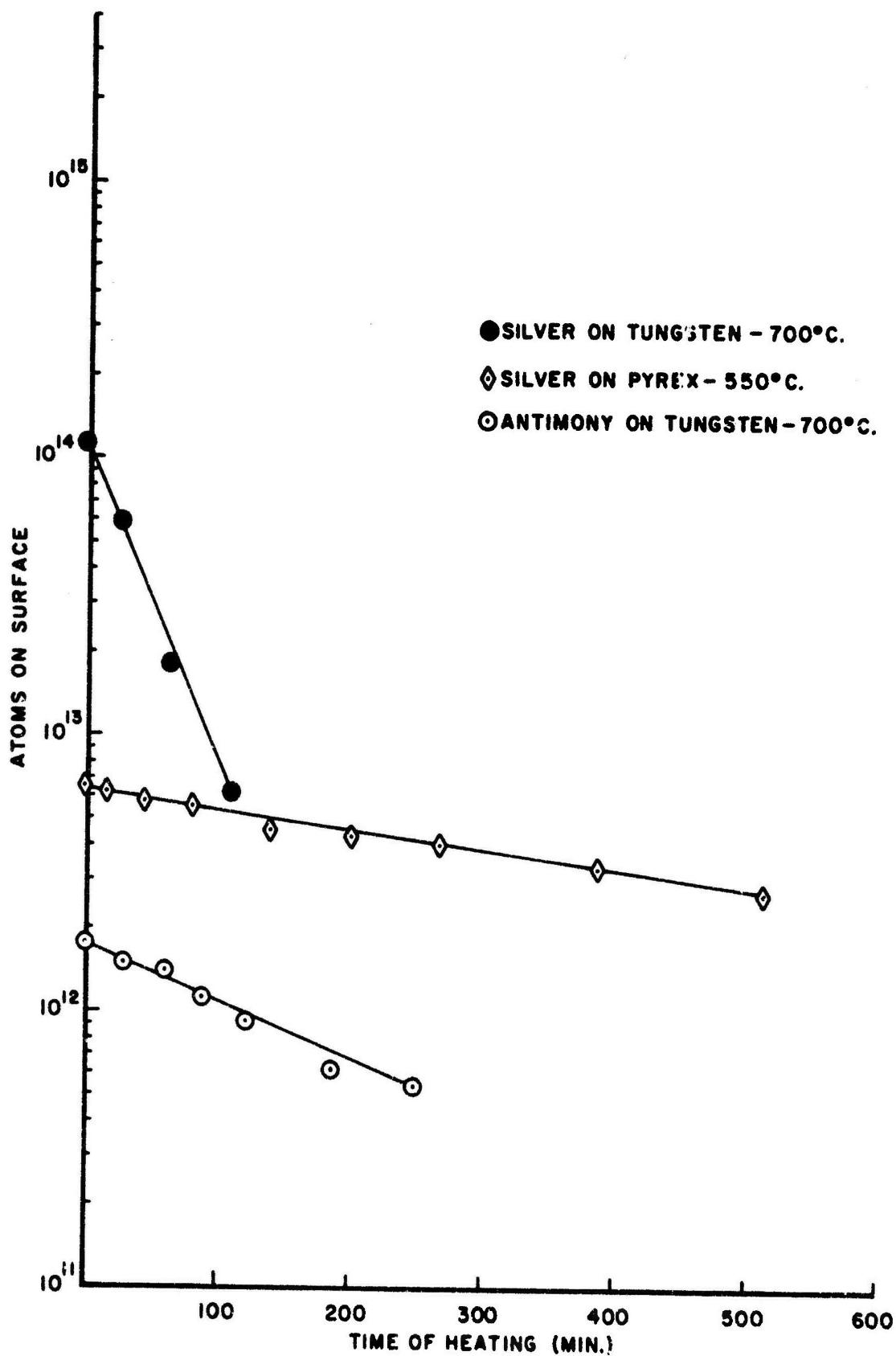


Figure 9 Silver and Antimony Desorption Curves.

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